# Combined Mass- or Volume-Fractal and Surface-Fractal Scattering Model

The model predicts $Q^{-Dv}$ scattering (i.e. between $Q^{-1}$ and $Q^{-3}$) for mass- or volume-fractals, and $Q^{-(6-Ds)}$ scattering (i.e. between $Q^{-3}$ and $Q^{-4}$) for surface-fractals. In the model function for $d\Sigma/d\Omega$ as a function of Q, there are four components:

$$d\Sigma/d\Omega = \{\text{VOLUME FRACTAL} + \text{SINGLE GLOBULE}\}\ \text{TERM} \\ + \text{SURFACE FRACTAL} + \text{FLAT BACKGROUND SCATTERING} \qquad [1]$$

These components are incorporated into the full theoretical expression as follows:

$$\frac{d\Sigma}{d\Omega} = \phi_{CSH} V_p \left|\Delta\rho\right|^2 \left\{ \frac{\eta R_C^3}{\beta R_o^3} \left(\frac{\xi_V}{R_C}\right)^{Dv} \frac{\sin\left[\left(D_V - 1\right)\arctan\left(Q\xi_V\right)\right]}{\left(D_V - 1\right) Q\xi_V \left[1 + \left(Q\xi_V\right)^2\right]^{(Dv-1)/2}} + \left(1-\eta\right)^2 \right\} F^2(Q)$$

$$+ \frac{\pi \xi_S^4 \left|\Delta\rho\right|^2 S_o \Gamma\left(5 - D_S\right) \sin\left[\left(3 - D_S\right)\arctan\left(Q\xi_S\right)\right]}{\left[1 + \left(Q\xi_S\right)^2\right]^{(5-Ds)/2} Q\xi_S} + \text{BACKGROUND} \qquad [2]$$

The first volume-fractal term contains $\phi_{CSH}$, $\xi_V$, and the mean radius, $R_o$, and shape aspect ratio, $\beta$, of the building-block C-S-H gel globules in the volume-fractal phase, here assumed to be spheroids. It also contains a local volume fraction, $\eta$, and the mean correlation-hole radius, $R_c$: the mean nearest-neighbor separation of the gel-globule centers. $R_c$, assumed to be weighted over spheroid surface-contacts, is given by:

$$R_C = \frac{R_o\sqrt{2}}{\chi_s} \left\{ 1 + \left(\frac{2+\beta^2}{3}\right) \chi_s^2 \right\}^{1/2} \qquad [3]$$

where:

$$\chi_s = (1/2\beta)\left\{1 + \left[\beta^2/\sqrt{1-\beta^2}\right]\ln\left(\left(1+\sqrt{1-\beta^2}\right)/\beta\right)\right\} \quad \text{for } \beta < 1, \qquad [4a]$$

and

$$\chi_s = (1/2\beta)\left\{1 + \left[\beta^2/\sqrt{\beta^2-1}\right]\arcsin\left(\sqrt{\beta^2-1}/\beta\right)\right\} \quad \text{for } \beta > 1 \qquad [4b]$$

In fitting the data, the need to incorporate $R_c$ with $\eta$, and a well-defined single-globule term (in addition to the volume-fractal) in the first bracket of eq. [1], is strong evidence for a solid volume-fractal phase. A well-defined single-globule term arises because, unlike the case of fractal pores in clays and porous rocks, nearest-neighbor solid particles cannot exist inside each other, i.e., their centers cannot approach, on average, to within $R_c$. This correlation-hole effect means that, for length-scales of order $R_o$, the individual particles are seen as distinct objects, even when incorporated into an aggregated structure. For a spheroid of aspect ratio, $\beta$, the form-factor for a single globule, $F^2(Q)$, is given by:

$$F^2(Q) = \frac{\pi}{2}|\Delta\rho|^2 V_p^2 \left| \int_0^1 \frac{J_{3/2}\left(QR_o\left[1 + (\beta^2-1)X^2\right]^{1/2}\right)}{\left(QR_o\left[1 + (\beta^2-1)X^2\right]^{1/2}\right)^{3/2}} dX \right|^2 \qquad [5]$$

where $V_p = (4\beta\pi R_o^3/3)$, $J_{3/2}(x)$ denotes a Bessel function of order 3/2, and X is an orientational parameter, here integrated over all orientations of the spheroid with respect to Q. Use of a mildly spheroidal globule shape avoids the pronounced Bessel function oscillations for spheres ($\beta = 1$), which can perturb the fit at high Q. Satisfactory fits are obtainable with both mildly oblate ($\beta = 0.5$) and mildly prolate ($\beta = 2$) aspect ratios, giving globule sizes equivalent to a 5 nm sphere for cement.

The surface fractal term in eq. [2] includes $\xi_S$, the mean upper limit of surface-fractal behavior at which the measured smooth surface area per unit sample volume is $S_o$. (The term, $\Gamma(5\text{-}Ds)$ is a mathematical gamma function.) The BACKGROUND term refers to the incoherent flat background scattering, and it is usually subtracted out of both data and fits for convenience.